
Characterization of Plutonium Oxides by Alpha-Induced Prompt Gamma-Ray Analysis

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Introduction

Legacy materials are segregated into broad material categories based on material origination and process history.

Item descriptions can be inadequate, incomplete, or simply wrong.

Chemical analysis of legacy materials is impractical.

A non-destructive matrix identification technique to identify or sort materials could prove useful.

- Long-term storage
- Materials disposition (MOX)

Materials Packaging/Stabilization

Guidance for long-term storage of PuO_2 specified by DOE Standard: DOE-STD-3013-2000.

Basic requirements:

- > 30 wt.% Pu + U
- Net weight < 5.0 kg (4.4 kg Pu)
- Stabilization at 950 °C for > 2 hours
- Require < 0.5 weight percent moisture

Risks Identified

- Container pressurization
- Container corrosion

Alpha-Induced Reactions

Alpha particles interact with matrix elements in (α,n) and (α,p) reactions.

Characteristic gamma-rays are produced that can be used to Matrix constituents.

Previous Work:

Giles, I. S. and M. Peisach, “*A survey of the analytical significance of prompt gamma-rays induced by 5 MeV alpha-particles,*” *Journal of Radioanalytical Chemistry*, Vol. 50, No. 1-2 (1979) 307-360.

Martin, H. Rodney, “*Reaction Gamma Rays in Plutonium Compounds, Mixtures and Alloys,*” Rocky Flats Publication, RFP-2832, 1975.

Tabulated Sensitivities

Order of magnitude sensitivity for elemental analysis using 5 MeV alpha particle bombardment.

< 1 %

Li, B, N, F, Na, Al, P, V, Mn, Rh

> 1 %, < 10 %

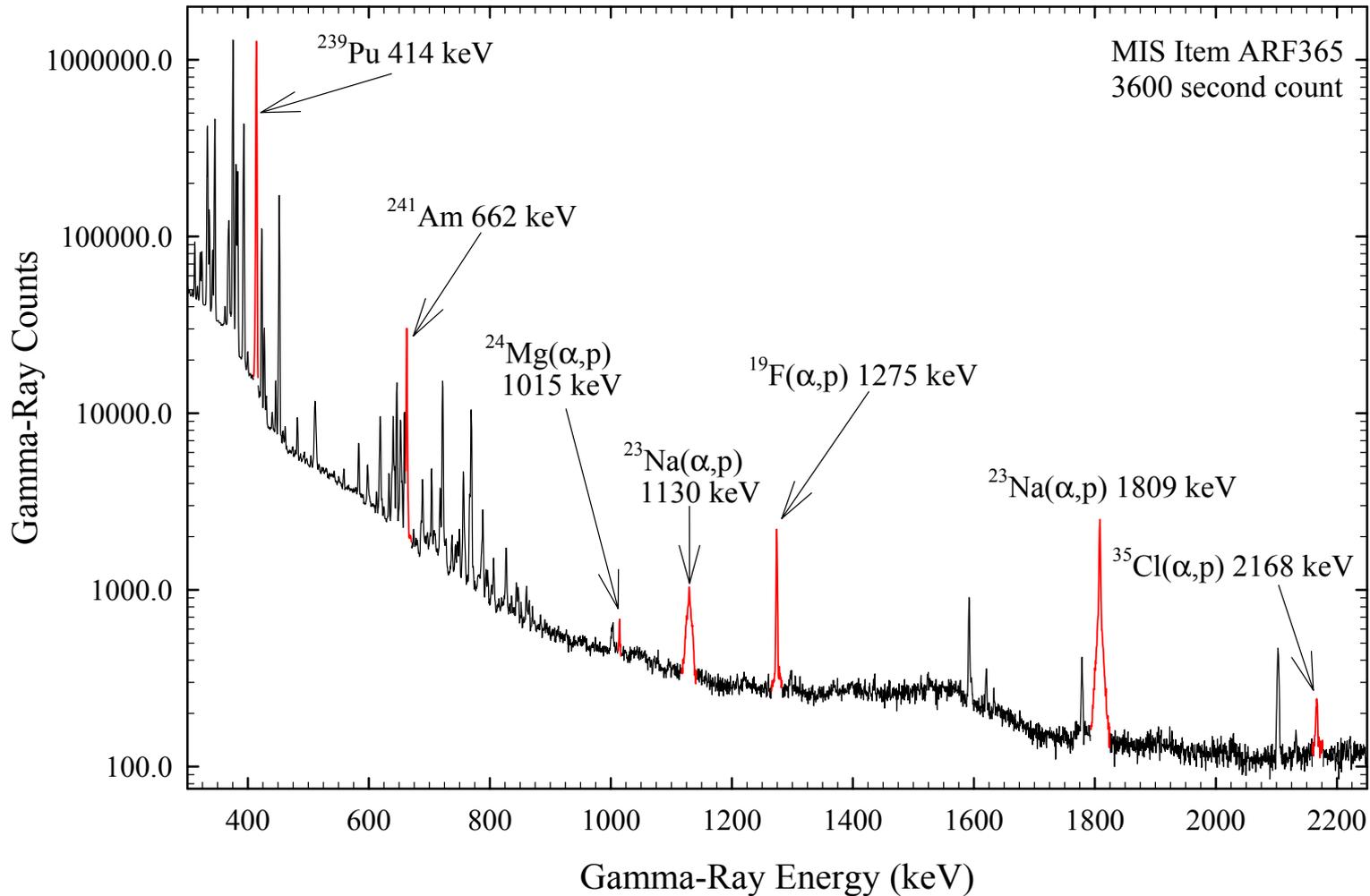
O, Mg, Si, Cl, K, Ti, Fe, Br, Mo, Ru,
Pd, Ag, Cd, Ta, W, Re, Ir, Pt, Au

> 10 %

Sc, Cr, Cu, Zn, Rb, Zr, Er, Hf, Hg

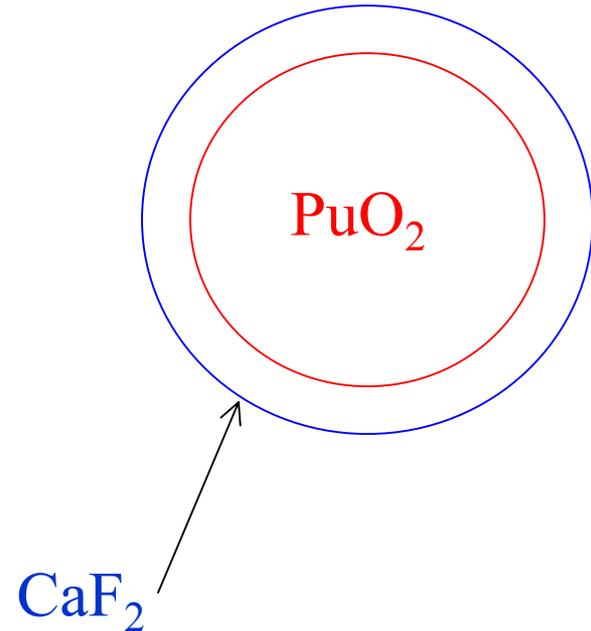
Journal of Radioanalytical Chemistry, Vol. 50 (1979) 307-360.

Prompt Gamma-Ray Spectrum

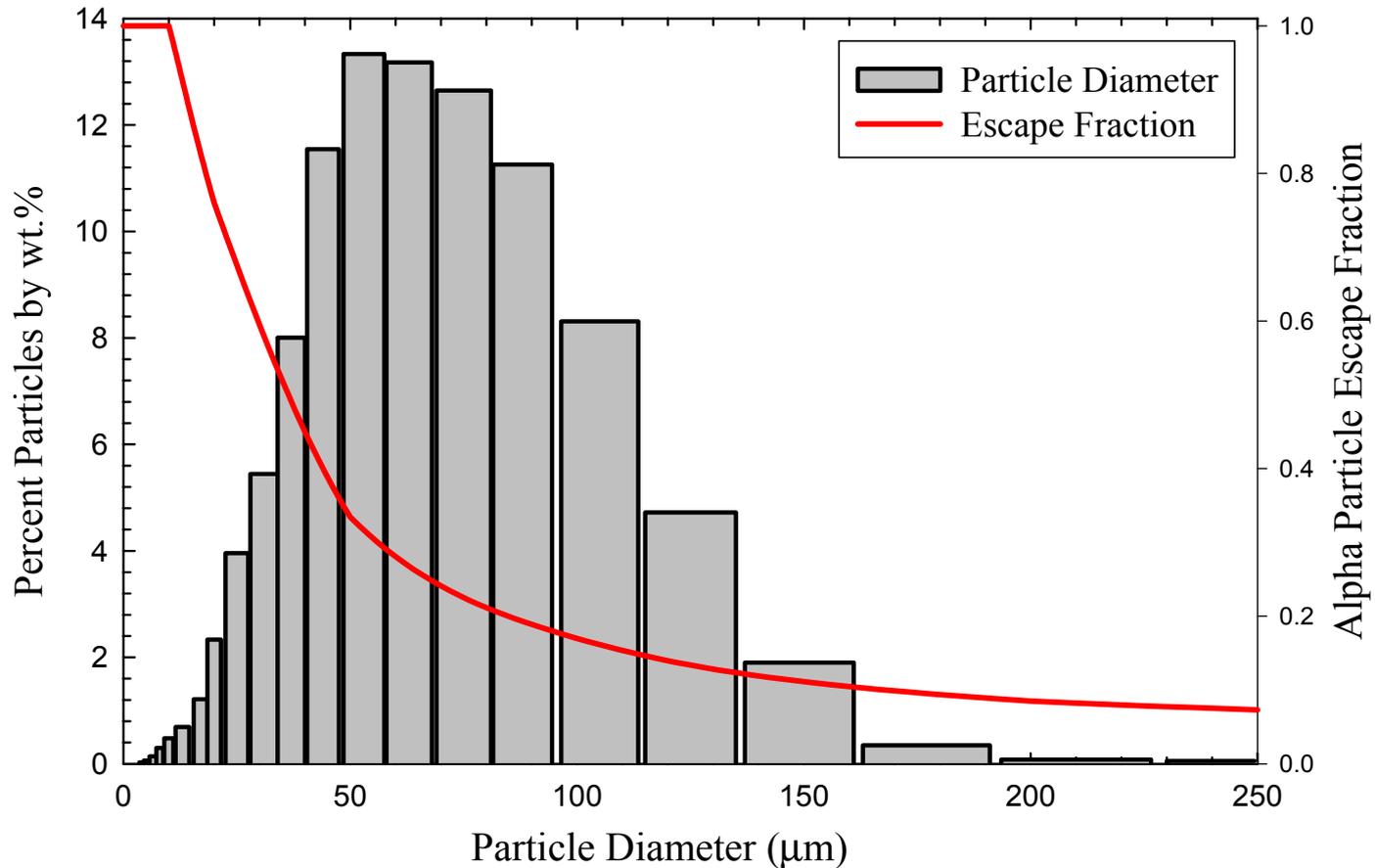


Reaction Rate Modeling

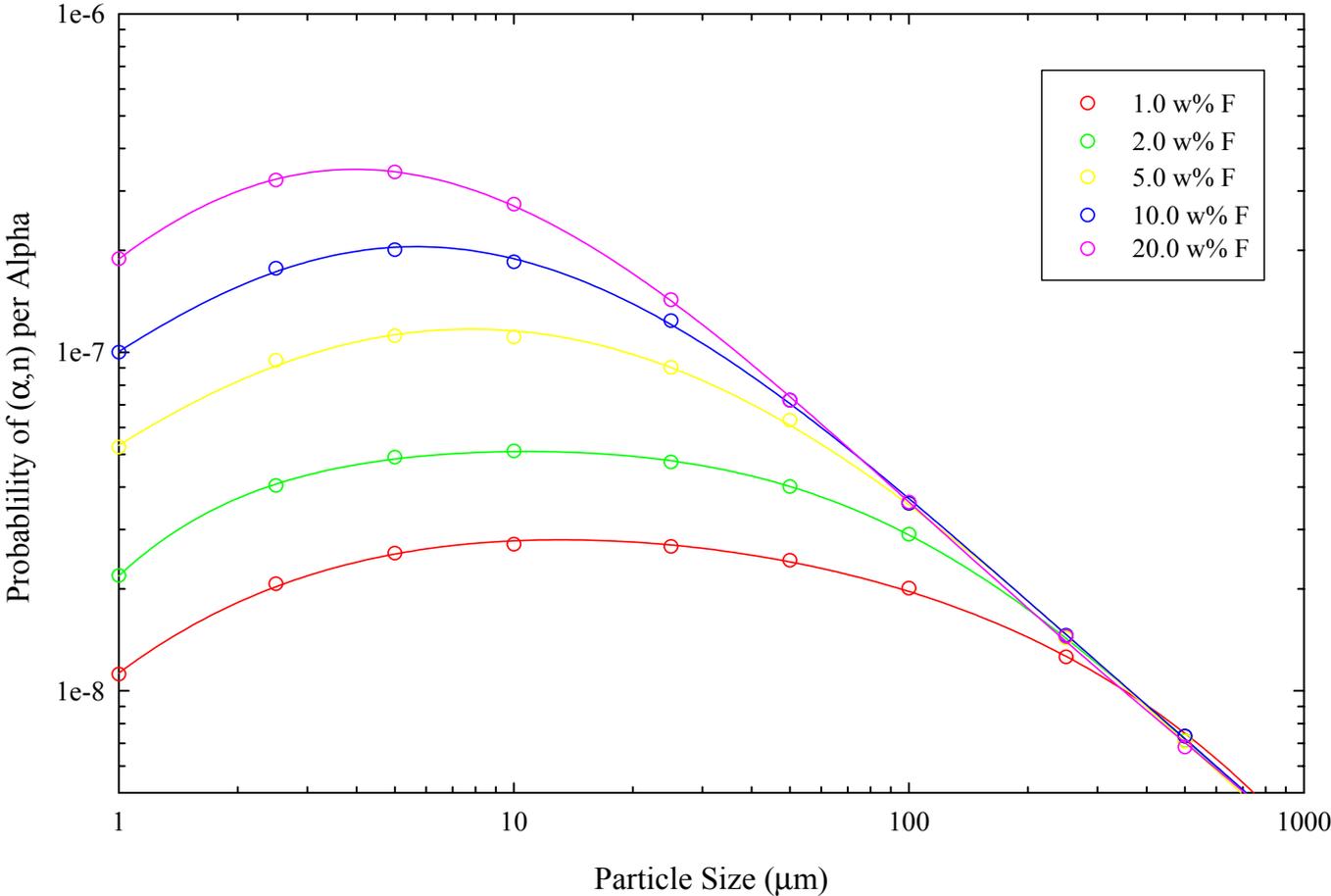
- Spherical PuO_2 particles
- Uniform coating of CaF_2
- Adjust thickness of CaF_2 to adjust weight fraction F
- 5 MeV alpha particles uniformly distributed throughout oxide



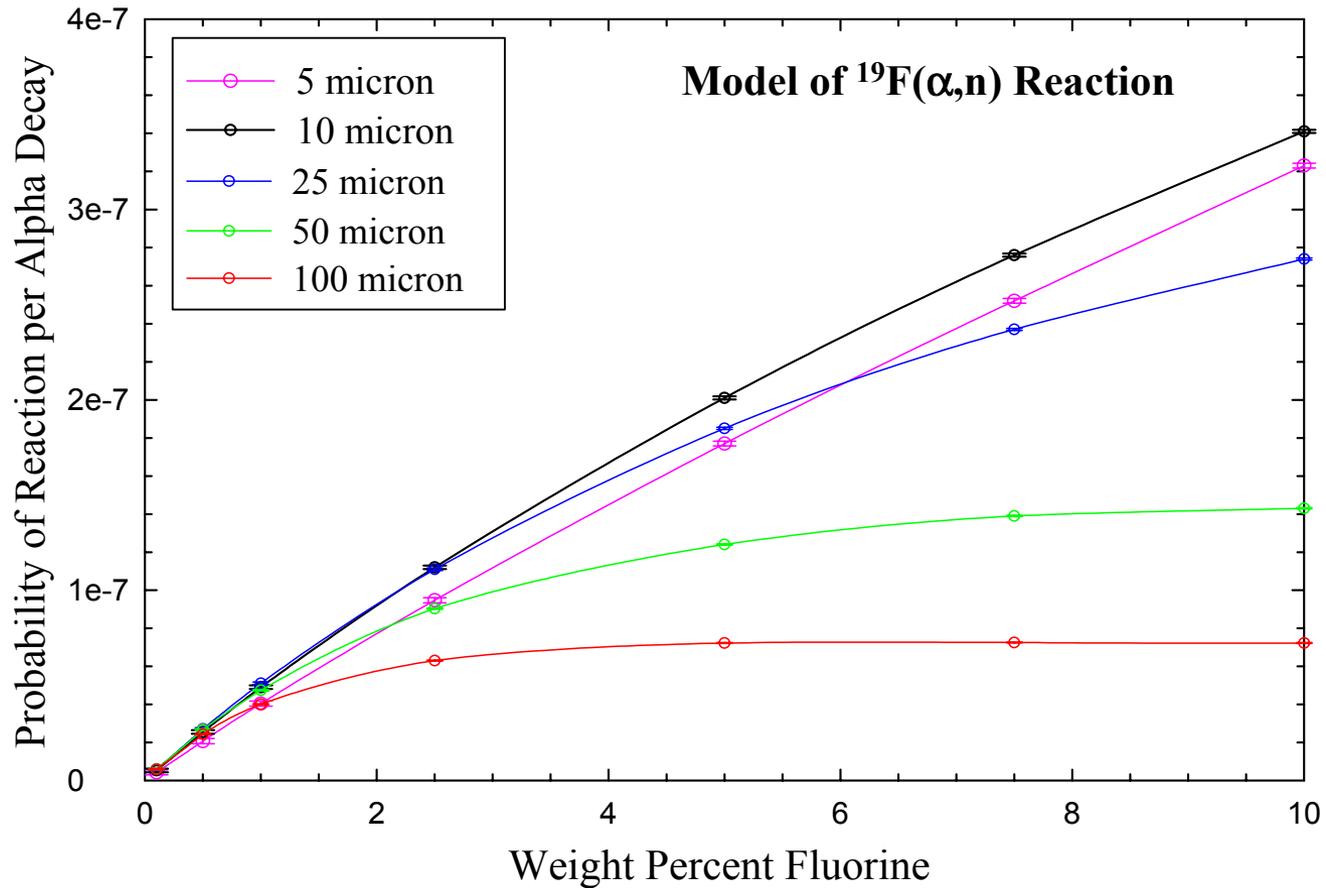
Particle Size and Escape Fraction



Interaction Probability



Reaction Rate Results



Salt Experiment

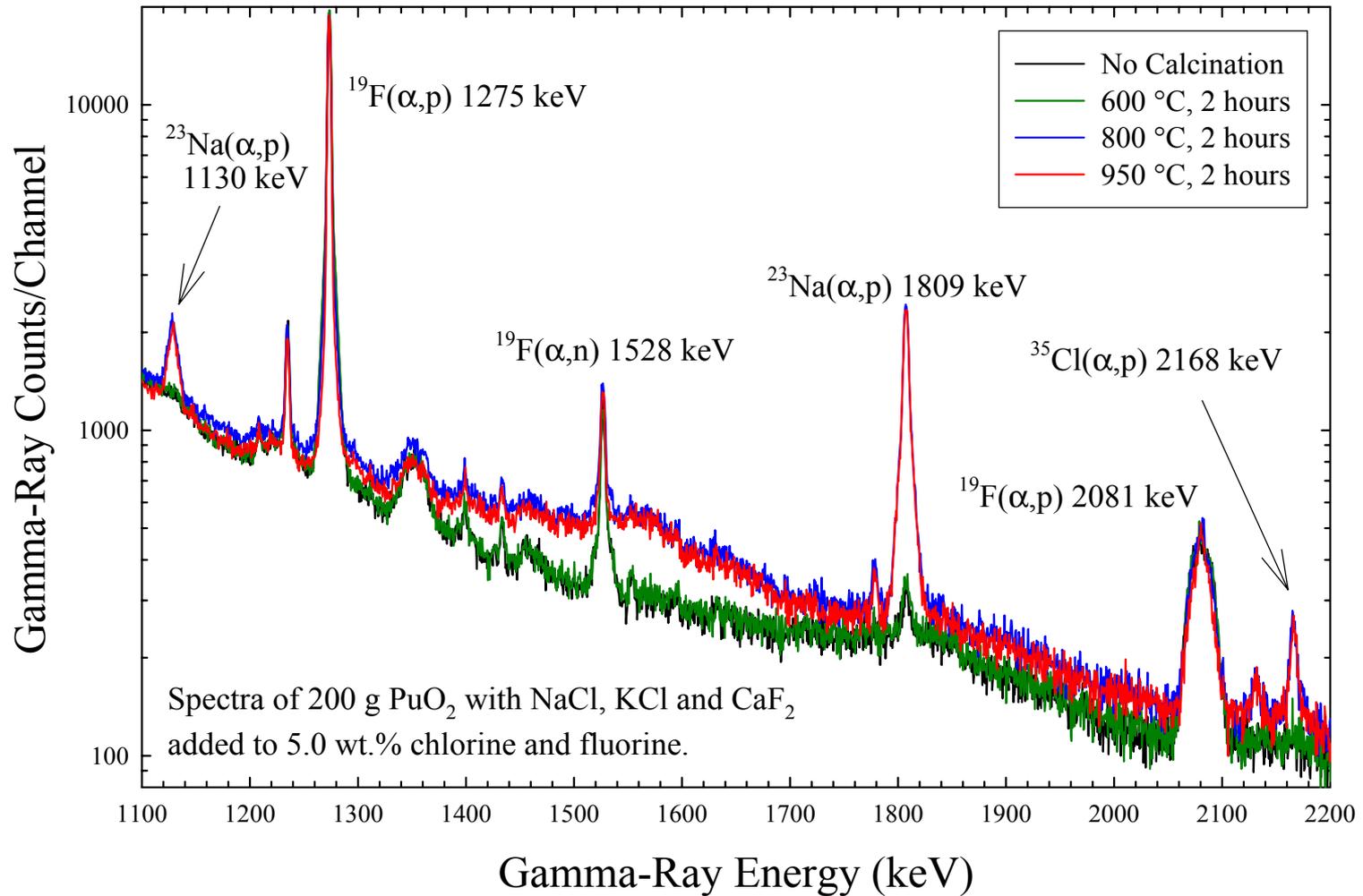
Add known amounts of salt to plutonium oxide

- 200 g pure PuO_2
- 50:50 NaCl-KCl mixture – 0.5, 5.0 and 20% Cl
- CaF_2 – 0.5, 5.0 and 10% F

Typical gamma-ray isotopics system used for measurements

- 20% Coaxial high-purity Ge detector
- Gamma-ray energy region of interest from 1 – 5 MeV

Effect of Calcination on Reaction Rates



Measurement MIS Materials

Plutonium bearing materials were sent to Los Alamos from Hanford and Rocky Flats.

Materials were characterized before and after stabilization to support 3013 disposition.

- Chemical analysis
- Physical properties
- Gas Analysis
- Moisture Measurements
- Non-Destructive Assay
- Prompt-Gamma Analysis (post-stabilization)

Calculations

Calculations for MIS Items: (Cl as an example.)

$$\textit{Normalized Count Rate} = \frac{A_{2168}}{LT \cdot \alpha}$$

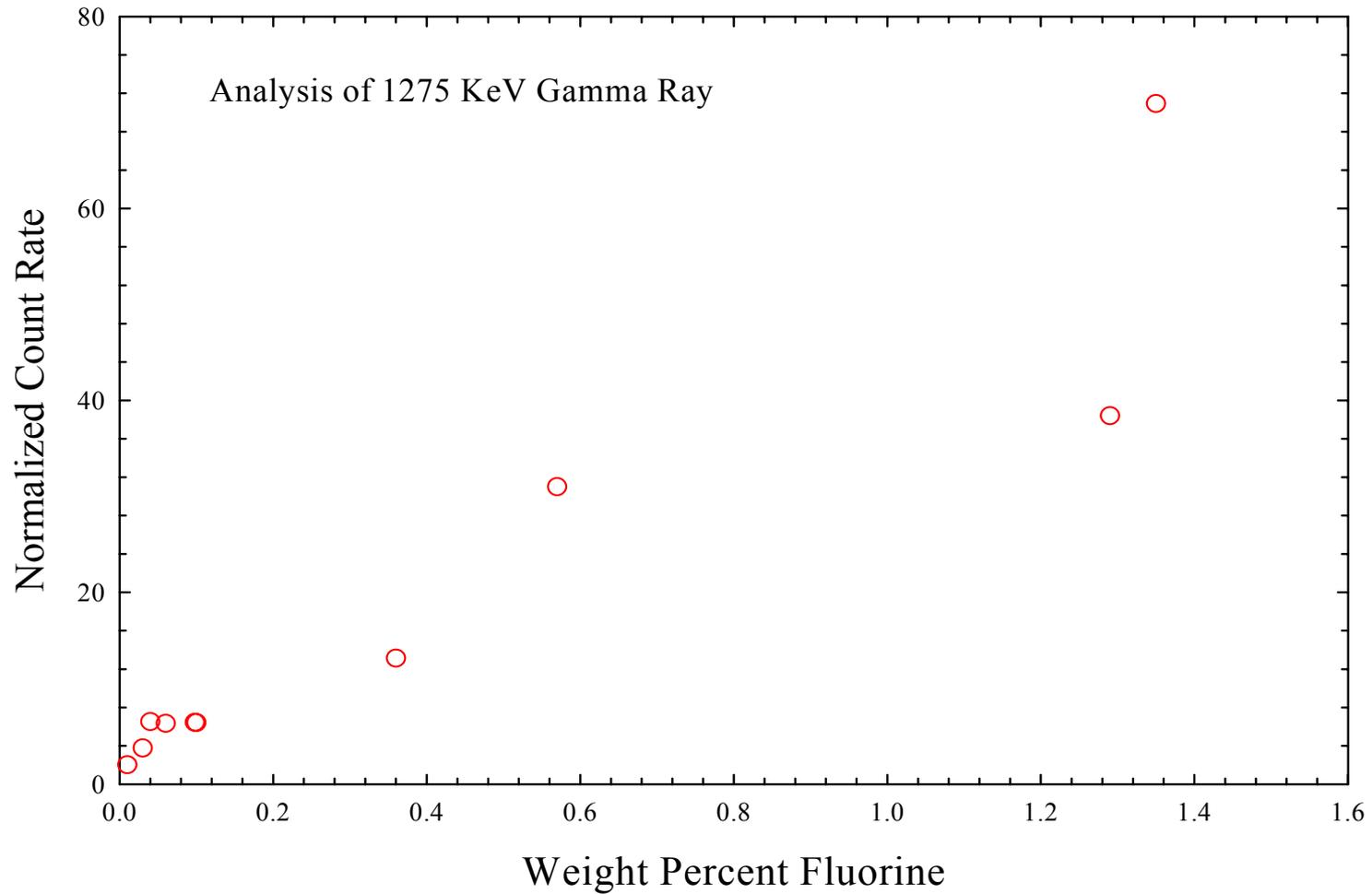
A_{2168} = Area under 2168 keV peak [$^{35}\text{Cl}(\alpha, p)$]

LT = MCA live time

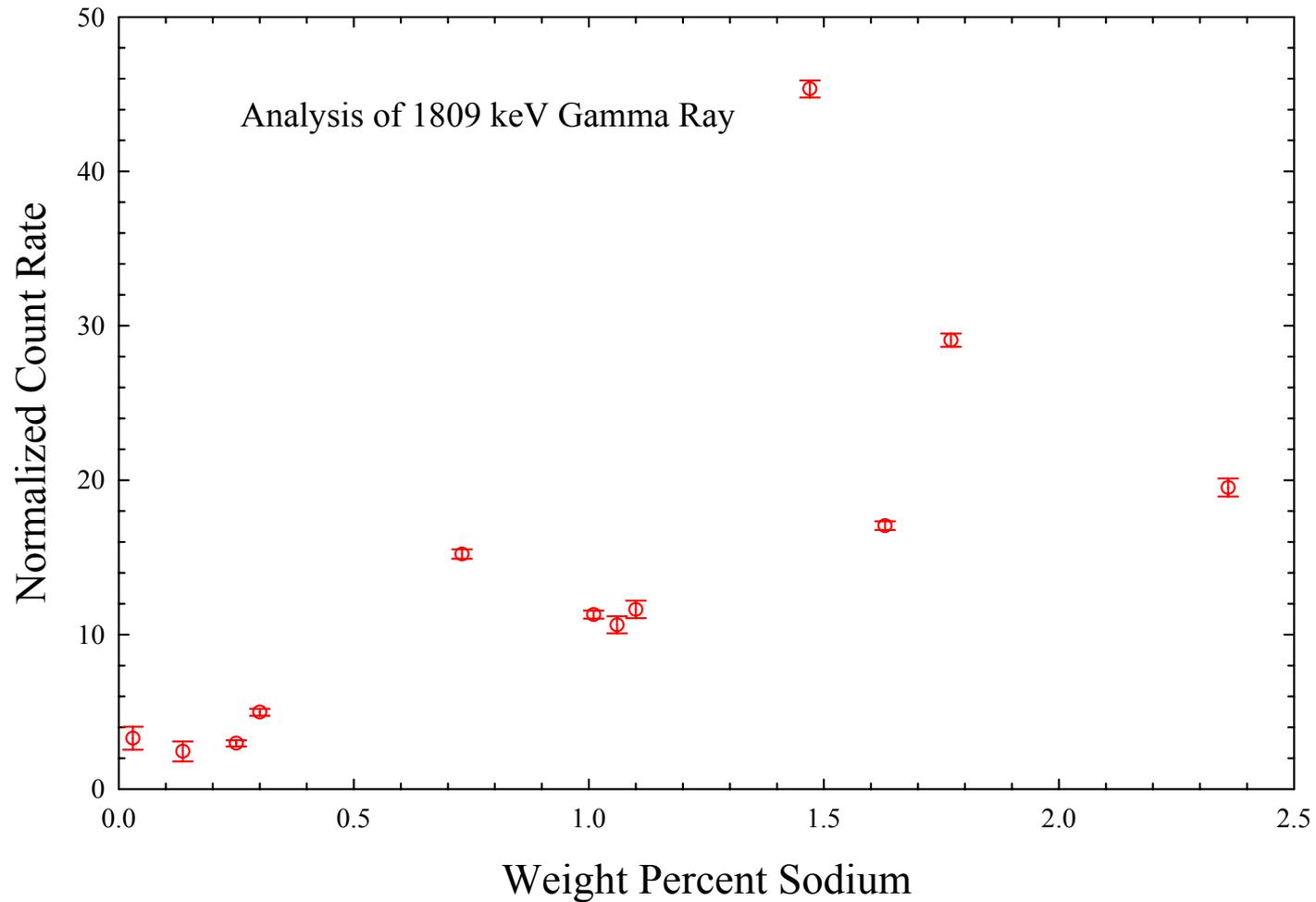
α = Alpha activity of sample (414 and 662 keV gamma-rays)

Items were analyzed for Cl, F, Na, Mg and Be using a simple region of interest analysis with linear background subtraction.

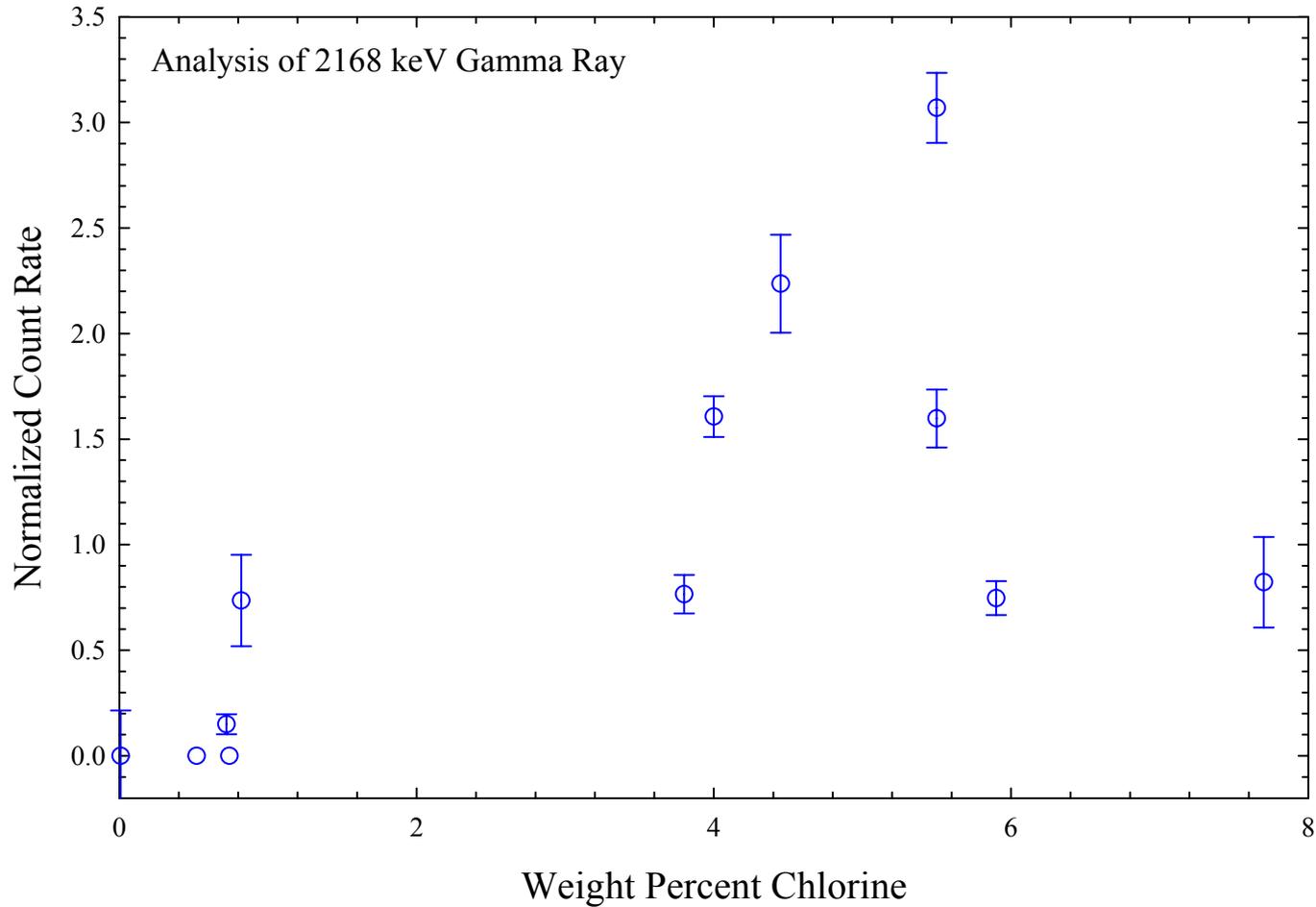
Prompt Gamma Analysis of Fluorine



Prompt Gamma Analysis of Sodium



Prompt Gamma Analysis of Chlorine



Detection Limits

Elemental detection limits in weight percent for several MIS Program items.

Parameters: 20% Ge detector, count time 3600 seconds.

	PuO₂	ARF365	MISNE4	ARF295	J2201
g Pu	3023 g	673 g	2754 g	128 g	472 g
Na	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %
F	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %
Mg	0.2 %	0.5 %	0.3 %	2.3 %	0.75 %
Cl	0.1 %	0.5 %	0.2 %	1.9 %	0.5 %
Be	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %	< 0.1 %

Conclusions

Successfully demonstrated identification of matrix constituents using alpha-induced interrogation.

Elements readily identified by prompt-gamma ray analysis of MIS items include: Cl, F, Na, Mg and Be.

Other elements identified include: Si, Al, and Li.

Quantification probably not realistic from an analytical analysis perspective: high - some - none.